U.S DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE ATTORNEY'S DOCKET NUMBER TRANSMITTAL LETTER TO THE UNITED STATES LP-1799 **DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371** INTERNATIONAL APPLICATION NO. | INTERNATIONAL FILING DATE August 11, 1998 August 9, 1999 PCT/EP99/05757 TITLE OF INVENTION UNSATURATED OLIGOPHENOL CYANATES APPLICANT(S) FOR DO/EO/US FEB 0 9 2001 Ulrich DAUM and Alessandro FALCHEIIU

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items

RADEMARY and other information 1. X This is a FIRST submission of items concerning a filing under 35 U S.C. 371 This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C 371. This express request to begin national examination procedures (35 U.S.C 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U S C 371(b) and PCT Articles 22 and 39(1)

4. X A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date 5 \underline{X} A copy of the international Application as filed (35 U S C 371(c)(2)) a __ is submitted herewith (required only if not transmitted by the International Bureau) b x has been transmitted by the International Bureau c __ is not required, as the application was filed in the United States Receiving Office (RO/US) 6 ___ A translation of the International Application into English (35 U S C 371(c)(2)) 7 __ Amendments to the claims of the International Application under PCT Article 19 (35 U S.C 371(c)(3)). a __ are transmitted herewith (required only if not transmitted by the International Bureau) b _ have been transmitted by the International Bureau have not been made; however, the time limit for making such amendments has NOT expired d. have not been made and will not be made. 8 __ A translation of the amendments to the claims under PCT Article 19 (35 U.S C 371(c)(3)) 9. X An oath or declaration of the inventor(s) (35 U.S C 371(c)(4)) 10 __ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U S.C 371(c)(5)) Items 11 to 16 below concern other document(s) or information included: 11 An Information Disclosure Statement under 37 C F R 1 97 and 1 98 12 X An assignment document for recording A separate cover sheet in compliance with 37 C F R 3 28 and 3 31 is included 13 X A FIRST preliminary amendment A SECOND or SUBSEQUENT preliminary amendment 14 __ A substitute specification. 15 __ A change of power of attorney and/or address letter 16 Other items or information

PCT Internationaler Vorläufiger Prüfungsbericht

**Please enter the Preliminary Amendment before calculating the filing fee. **

U.S. Application NO (If know		TIONAL APPLICATION NO		LP-1799	MBER
$\frac{1}{\sqrt{109}}$		/EP99/05757		CALCULATIONS	PTO USE ONLY
17 X The following to Basic Nation	al Fee (37 C.F.R. 1.492(a)(1)-(5):	_	4 000 00		
	been prepared by the EPO or JPC		1,000.00	\$1,000.00	
	inary examination fee paid to USP		\$720 00		
No international pre international search	eliminary examination fee paid to U h fee paid to USPTO (37 C F.R 1.	JSPTO (37 C F R 1 482) 445(a)(2))	\$790 00		
Neither internations search fee (37 C.F.	al preliminary examination fee (37 .R 1.445(a)(2)) paid to USPTO	C F R 1 482) nor interna	tional \$1070 00		
International prelim	ninary examination fee paid to USP ovisions of PCT Article 33(2)-(4)	PTO (37 C F R. 1 482) an	d all \$98.00		
	NTER APPROPRIA	TE FEE AMO	UNT	\$1,000.00	
Surcharge of \$130 0	0 for furnishing the oath or declara	tion later than 20		\$	
Claims	lest claimed priority date (37 C.F F	Number Extra	Rate		
Total Claims	-20 =		X \$18 00	\$	
Independent Claims	-3 =		X \$78 00	\$	
Multiple Dependent	Claim(s) (if applicable)		X \$270 00	\$	
		F ABOVE CALCULATIO	NS =	\$1,000.00	
Reduction by 1/4 for f	iling by small entity, if applicable			\$ 1,000.00	
	Note 37 C.F.R 1.9, 1.27, 1.28)	SUBTOT			"
				\$1,000.00	
Processing fee of \$1 months from the ear	30.00 for furnishing the English tra liest claimed priority date (37 C.F.I	anslations later than 2 R 1.492(f))	0 — 30	\$	
777071011011111111111111111111111111111		TOTAL NATIONAL	FEE =	\$1,000.00	
Fee for recording the	e enclosed assignment (37 C.F R	1 21(h)) The assignmen	nt must be	\$40.00	
accompanied by an	appropriate cover sheet (37 C F.R	. 3.28, 3 31) \$40 00 per TOTAL FEES ENCLO	property + SED =	\$1,040.00	
			· -	Amount to be:	\$
				Charged	\$
		<u> </u>		<u> </u>	1 7
a X A check in the	amount of \$1,040.00 to cover to	he above fees is enclose	d.		
b Please charge of this sheet is	my Deposit Account No	in the amount o	of \$	to cover the above	fees. A duplicate copy
c X The Commiss Account No. 0	ioner is hereby authorized to charg <u>6-1110</u> . A duplicate copy of this s	ge any additional fees wh heet is enclosed	ich may be	required, or credit any ove	erpayment to Deposit
NOTE: Where an a (b)) must be filed a	appropriate time limit under 37 C and granted to restore the applic	C.F.R. 1.494 or 1.495 has ation to pending status	not been i	met, a petition to revive ((37 C.F.R. 1.137(a) or
			1	11 1/4 0	
SEND ALL CORRE	SPONDENCE TO	SI	SNAT RE	H. Merch	
Virgil H. Marsh Fisher, Christen &		Si			
Suite 1401, 1725 K Washington, DC 2			.,	t	
		1	Virgil H. N IAME	Marsh	
Tel.: 202/659-2000 February 9, 2001					
, , , , , , , , , , , , , , , , , , , ,			23.083		
		_	REGISTRATIO	N NUMBER	

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Docket: LP-1799

Applicant

DAUM, Ulrich, et al.

Serial No.

Not yet assigned

[National Stage Based Upon

PCT/EP99/05757]

Filed

February 9, 2001

For

UNSATURATED OLIGOPHENOL CYANATES

PRELIMINARY AMENDMENT

Assistant Commissioner for Patents Washington, D.C. 20231

Sir:

Please amend this application as follows:

IN THE SPECIFICATION:

On page 1, between the title and the first line of the specification, insert the following:

-- This application is a continuation (national stage) application of International (PCT) Patent Application No. PCT/EP99/05757, filed on August 9, 1999, which is a continuing application of U.S. Provisional Application Serial No. 60/096,253, filed on August 12, 1998.--

REMARKS

The continuation/continuing application history of the application has been inserted on page 1 of the application.

Respectfully submitted,

Date: Jan. 9, 2001

Virgil H. Marsh Reg. No. 23,083

Fisher, Christen & Sabol Suite 1401 1725 K Street, N.W. Washington, D.C. 20006

Tel.: (202) 659-2000

L.P. 1799, PCT

LONZA AG, Münchensteinerstrasse 38, CH-4052 Basel

Unsaturated oligophenol cyanates

Unsaturated oligophenol cyanates

The Invention relates to oligophenol cyanates of the general formula

$$[A-]_n[B-A-]_xB[-A]_m$$
 (I).

In this formula, A is in each case a group of the formula

and B is in each case a group of the formula

15

 ${\ensuremath{\mbox{R}}}^1$, ${\ensuremath{\mbox{R}}}^2$ and ${\ensuremath{\mbox{R}}}^3$ on each group A independently of the others are in each case hydrogen or a bond to a group B with the proviso that each group A has either one or two bonds to B.

Both R^4 and $R^{4'}$, and R^5 and $R^{5'}$, on each group B independently of the others, are in each case either together a direct bond or, in any desired order, are hydrogen and a bond to a group A with the proviso that each group B has either one or two bonds to A. The indices m and n are 0 or 1 and x is an integer from 0 to 10 with the proviso that at least one of the numbers m, n and x is other than 0 and m and n are not both at the same time 1.

The Invention also relates in particular to mixtures of such compounds with one another and/or with those compounds of the

10

5

15

20

25

formula I in which m and n deviate from the above definitions by both being 1.

Saturated oligophenol cyanates of general formula I in which m and n deviate from the compounds of the present invention by both being 1 are known, for example, from EP-A-0 147 548 and are marketed by the Dow Chemical Co. under the designation XU71787. These compounds do not have any olefinic double bonds and are therefore able to polymerize only by cyclotrimerization of the cyanate groups or by reaction with functional groups of other compounds. The cyclotrimerization requires the presence of catalysts and/or high temperatures. In contrast it is often desirable to achieve partial curing or crosslinking by means, for example, of irradiation at room temperature. In addition, these known compounds have a relatively high viscosity, which is unfavourable for some applications.

The object of the present invention was therefore to provide oligophenol cyanates which have a low viscosity and which without further additives can be (partially) polymerized or crosslinked at room temperature by means, for example of radiation-induced free-radical reactions.

In accordance with the invention this object is achieved by the unsaturated oligophenol cyanates of the formula I in accordance with Claim 1. The molecule of these compounds has at least one olefinic double bond $(R^4-R^4'$ and/or R^5-R^5' according to formula I) which permits free-radical addition polymerization.

The olefinic double bonds are present in groups of the formula

30 The degree of polymerization, x, lies preferably between 0 and 5 and, with particular preference, between 0 and 3.

The unsaturated oligophenol cyanates of the invention can be prepared by reacting an oligophenol of the general formula

20

25

30

5

$$[A'-]_n[B-A'-]_xB[-A']_m$$
 (II),

in which A' is a group of the formula

$$\mathbb{R}^3$$
 \mathbb{R}^1

and B, R^1 , R^2 , R^3 , R^4 , R^4 , R^5 , R^5 , m, n and x are as defined in Claim 1, is reacted with cyanogen chloride in the presence of a tertiary amine. Oligophenols of formula II are obtainable from Borden Chemical Inc. under the designations ESD-X1 to -X5, ESD-472C and ESD-473C. The compounds concerned here are condensation products of dicyclopentadiene (dimeric cyclopentadiene) and phenol, which are present as a mixture of isomeric and/or homologous compounds and also contain fractions of saturated compounds where m = n = 1.

The preparation of the oligophenol cyanates of the invention is preferably carried out at a temperature of less than 10 $^{\circ}$ C in a polar solvent such as butyl acetate and/or acetone or methyl ethyl ketone, for example, or in mixtures of these solvents. Particularly preferred reaction temperatures are below 0 $^{\circ}$ C - for example, -10 $^{\circ}$ C. It is advantageous to employ 1.0-1.1 mol of tertiary amine and 1.0-1.2 mol of cyanogen chloride per OH equivalent of the oligophenol II. A particularly preferred tertiary amine is triethylamine.

The unsaturated oligophenol cyanates of the invention have a low viscosity at processing temperature and produce polytriazine resins having a particularly low dielectric constant. They are particularly suitable, for example, as matrix material for the production of fibre-reinforced composites, especially for

components in aerospace technology, or as base materials for the production of printed circuit boards. Owing to their low viscosity and capacity for polymerization by means of high-energy radiation (UV, X-rays, γ -rays or electron beams), they are also suitable for (photo)lithographic varnishes, solder resists for circuit boards, or other radiation-curable lacquers and coatings.

The following example illustrates the preparation of the oligophenol cyanates of the invention without constituting any restriction.

Example

Oligophenol ESD-X3 (Borden Chemical Inc.) was dissolved in n-butyl acetate/acetone (v/v = 80:20) to give a 15% strength solution. The solution was cooled to -10 °C and, at this temperature, 105% of the calculated amount of triethylamine and then, over the course of 30 minutes, 110% of the calculated amount of cyanogen chloride were added. After a further 30 minutes of reaction, the mixture was subjected to extraction with water, twice at 30 °C, in order to remove the ammonium salts formed, and was then passed twice through a falling-film evaporator in order to remove the solvent and the by-product, N, N-diethylcyanamide.

Yield: about 100%

25 Properties:

viscosity: 165 mPa·s (at 125°C)

degree of reaction: (phenol → cyanate) >98%

gel time: 25 min (at 200°C)

carbamates: <1%

30 N, N-diethylcyanamide: <2000 ppm

20

25

Claims

1. Unsaturated oligophenol cyanates of the formula

5
$$[A-]_n[B-A-]_xB[-A]_m$$
 (I),

in which A is in each case a group of the formula

and B is in each case a group of the formula

wherein R^1 , R^2 and R^3 are in each case hydrogen or a bond to a group B with the proviso that each group A has either one or two bonds to B;

both R^4 and R^4 , and R^5 and R^5 are in each case either together a direct bond or are hydrogen and a bond to a group A with the proviso that each group B has either one or two bonds to A; the indices m and n are 0 or 1 and x is an integer from 0 to 10 with the proviso that at least one of the numbers m, n and x is other than 0 and x and x are not both at the same time 1, and mixtures thereof with one another and/or with those compounds of the formula I in which x and x deviate from the above definitions by both being 1.

5

- 2. Oligophenol cyanates according to Claim 1, characterized in that x is from 0 to 5.
- 3. Process for preparing unsaturated oligophenol cyanates according to Claim 1, characterized in that an oligophenol of the general formula

$$[A'-]_n[B-A'-]_xB[-A']_m$$
 (II),

in which A' is a group of the formula

$$R^3$$
 R^1
 R^2

and B, R^1 , R^2 , R^3 , R^4 , R^4 , R^5 , R^5 , m, n and x are as defined in Claim 1, is reacted with cyanogen chloride in the presence of a tertiary amine.

- 4. Use of the unsaturated oligophenol cyanates according to Claim 1 as matrix material for fibre-reinforced composites.
- 5. Use of the unsaturated oligophenol cyanates according to Claim 1 as radiation-curable varnishes, resists, lacquers and coatings.

20

25

Abstract

5

The invention relates to unsaturated oligophenol cyanates of the general formula (I) $[A-]_n[B-A-]_xB[-A]_m$ in which A is a group of formula

and B is a group of formula

$$R^{5}$$
 R^{4}
 R^{4}

where R^1 , R^2 and R^3 are each hydrogen or a bond with a group B, there being either one or two bonds with B; and both R^4 and R^4 as well as R^5 and R^5 separately or jointly represent a direct bond or hydrogen and a bond with a group A, there being either one or two bonds with A. The indices m and n are 0 or 1 but not both 1 at the same time and x is a whole number between 0 and 10, where at least one of the numbers m, n and x is not 0.

The above unsaturated oligophenol cyanates can be prepared by reacting the corresponding oligophenols with cyanogen chloride. They have a low viscosity and owing to their double bonds are able to undergo free-radical polymerization. They are especially suitable for use as matrix materials for fibre-reinforced composites and for radiation-curable varnishes and coatings.

Type a plus sign (+) ins	U.S. Department of Com	+ Democroe		
Rev 6/95	Patent and Trademark (Attorney Docket Number	r LP-1799
			First Named Inventor	DAUM
DEC	LARATION		COMPL	ETE IF KNOWN
			Application Number	
X Declaration	OR Declara	tion	Filing Date	
Submitted with Initial Filing	Submitte Initial Fi		Group Art Unit	
			Examiner Name	
As a below named inventor, I he	reby declare that:	<u>L</u>		1 · · · · · · · · · · · · · · · · · · ·
l believe I am the original, first ar are listed below) of the subject n	natter which is claimed a	and for which a patent is		r (if plural names
the specification of which	(Title of ti	he Invention)		
x is attached hereto				
OR was filed on (MM/DD	ommo [as United States Application Numb	er or PCT International Application
Number		amended on (MM/DD/Y		(if applicable)
I hereby state that I have review	ed and understand the c	ontents of the above ide	entified specification, including the cla	
specifically referred to above.				
acknowledge the duty to disclos	se information which is r	naterial to patentability	as defined in Title 37 Code of Federal	Regulations, § .56.
certificate, or §365(a) of any PC	T international application secking the box, any fore	n which designated at le eign application for pater	a)-(d) or §365(b) of any foreign applic east one country other than the United nt or inventor's certificate, or of any Po	d States of America, listed below and
Prior Foreign Application Number(s)	Country	Foreign Filing Date (MM/DD/YYYY)	Priority Not Claimed	Copy Attached? YES NO
PCT/EP99/09477	International	08/09/1999		\square
98202692.4	Europe	08/11/1998		x
60/096,523	U.S. Provision	al 08/12/1998	x	
Additional foreign ap	plication numbers are lis	sted on a supplemental	priority sheet attached hereto:	
I hereby claim the benefit under	Title 35, United States 0	Code §119(e) of any Uni	ted States provisional application(s) li	isted below.
Application Number(s)	Filing C	Date (MM/DD/YYYY)	Additional provisional
·				application numbers are listed on a supplemental priorit sheet attached hereto.

DECLARATION	Page 2				

I hereby claim the benefit under Title 35, United States Code §120 of any United States application(s), or §365(c) of any PCT international application designating the United States of America, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT international application in the manner provided by the first paragraph of Title 35, United States Code §112, I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations 1 56 which became available between the filing date of the prior application and the national or PCT filing date of this application

PCT Parent Number	Parent Filing Date (MM/DD/YYYY)	Parent Patent Number (if applicable)
PCT/EP99/05757	08/09/1999 08/12/1998	
	Number	Number (MM/DD/YYYY) 08/09/1999

Additional U.S. or PCT international application numbers are listed on a supplemental priority sheet attached hereto

As a named inventor, I hereby appoint Trademark Office connected therewith	nt the following attorney(s) and/or agent(s) t th	o prosecute this application and to transa	ct all business in the Patent and
Firm Name Fisher, Ch	risten & Sabol	Payor Number (if applicable)	
Name	Registration Number	Name	Registration Number
Virgil H. Marsh	23,083_		
Kara M. Armstrong	38,234		
Additional attorney(s) and/or agent(s) named on a supplen	nental sheet attached hereto	

x Please direct all correspondence to	Name	Virgil H. Marsh			
Address Fisher Christen &	Sabol				
Address Suite 1401, 1725 K	Street,	N.W.			
city Washington		State D.C.	Zip	20006	
country USA		Telephone (202)659-2000	Fax	(202)659-2015	
I hereby declare that all statements made her	roin of my own	knowledge are true and that all statements made	to on infor	mation and haliof are helio	_

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true, and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon

Nam	e of Sole or First Inve	ntor:		A petition has b	een filed for this unsign	ed inventor	····	7
Given Name	Ulrich	Middle Initial	Fami Name		M	Suffix		7
Invent Signat		2.	du	-		Date	01/26	01
Residen City	ce: Hofstetten	Co	^{untry} Switz	erland		Citizenship		7', f
Post Off	ice Address: Chöpfliw	eg 17	. 1			<u> </u>		7 67
City	Hofstetten	Postal Code	CH-4114	Country	Switzerland	Appli Autho		1
X	Additional inventors are being	g named on supplem	nental sheet(s) attac	ched hereto				



Type a plus	s sign (+) inside th	nis box	\rightarrow	+					
	DECLARA1							INVENTOF ntal Sheet	R(S)
Name of Ad	lditional Joint Inve	ntor, if	any	':		petition has be	en filed for this ur	isigned inventor	
Given Ale		Middle Initial 1			Family Name	FALC	HETTO	Suffix	
Inventor's Signature	100	a	1	11	Name			Date	
<u> </u>	H Growng H	, <u> </u>	<u>W</u>		0			31,01.01	<u> </u>
Residence: DO	modossola	1	Cour	itry	Italy			Citizenship	Italy
Post Office Addres	ss: Via Leopard	i 2/c							
city Domo	odossola	Pos		1-288	345	Country	Italy	Applica Authorit	
Name of Ad	lditional Joint Inve	ntor, if	any	':		petition has be	en filed for this ur		
Given		Middle			Family			Suffix	
Name Inventor's		Initial			Name	<u> </u>		Date	
Signature									
Residence: City			Cour	itry				Citizenship	'
Post Office Addres	;s: 								
City		Cod				Country		Applica Authori	
Name of Ad	lditional Joint Inve	entor, if	fany	' :	□ A	petition has be	een filed for this ur	signed inventor	
Given Name		Middle Initial			Family Name			Suffix	T
Inventor's Signature		miniai			INdille			Date	
Residence:			Cour	ster. I				Citizenship	
City Post Office Addres			Cour	ilry				Citizenship	<u>'</u>
City		I Boo	stal			Country		Applica	nt I
		Cod	de			Codinary		Authori	
	lditional Joint Inve		rany	/: 	1	petition has be	een filed for this ur		
Given Name		Middle Initial			Family Name			Suffix	
Inventor's Signature								Date	
Residence:	. =		Cou	ntry				Citizenship	,
City Post Office Addres	ss:								İ
City		Pos	stal			Country		Applica	nt
Name of Ac	ditional Joint Inve	Contor it		,.	T =			Authori	ty
		Middle	,	•		petition has b	een filed for this ur		,
Given Name Inventor's		Initial			Family Name			Suffix	
Signature								Date	
Residence:			Cou	ntry				Citizenship	5
City Post Office Addres	ss:			i_					
City			stal			Country		Applica	
Name of Ac	dditional Joint Inve	Co entor. if		/: /:				Authori	ty
Given						petition has b	een filed for this ur	nsigned inventor	
Name Inventor's		Initial			Family Name			Date	
Signature								Dute	
Residence: City			Cou	ntry				Citizenship	,
Post Office Addres	ss:								1
City			stal			Country		Applica	
L		Co	de			L	1	Authori	ty

ITX